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EVOLUTION OF POTTERY PRODUCTION DURING LATE NEOLITHIC PERIOD AT SIALK ON KASHAN PLAIN, CENTRAL PLATEAU OF IRAN

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The prehistoric sherds recovered from the North Mound of Tepe Sialk, were investigated using XRF, XRD and SEM/EDX analyses. These studies showed the occurrence of a gradual evolution in pottery making from the Sialk I to Sialk II periods eventually leading to the production of bulk red pottery at the final phase of Sialk II. The relative similarity of compositions, homogeneous microstructures and the presence of high-temperature phases demonstrated a high degree of specialisation in the selection of raw materials and control of the firing temperature and atmosphere among the potters of Sialk in the sixth millennia BC, peaking at the final phase of Sialk II.

KEYWORDS: SIALK, CENTRAL PLATEAU OF IRAN, NEOLITHIC POTTERY, XRD, MICROSTRUCTURE, XRF

INTRODUCTION

The Central Plateau of Iran is one of the most important regions for studying the prehistory of this country. The societies of this region have been at the centre of at least three millennia of sustained and continuous change and development from the sixth millennia BC onward playing an active role in cultural and technical-economic development through their intraregional and interregional interactions. The deep deposits of archaeological remains, reaching to ~16 meters in some sites, along with the sustained progress and advancement in technology and innovation make this region very attractive for the studies of prehistoric regions of Iran.

The systematic Archaeological research in the Central Plateau began in 1931 with Erich Schmidt's excavation of Tepe Hissar (Schmidt 1937). Afterwards many archaeologists have also been engaged in the study of historical, cultural, technological and socio-political development of the Central Plateau and a number of chronological models have been proposed. In the earlier studies of the region the culture-historical viewpoint was mainly predominated and the scholars focused heavily on stylistic changes in ceramics.

For example, McCown in his paper entitled 'The comparative stratigraphy and chronology of Iran' (McCown 1954) provided the first synthesis of the relationships

among the early cultures of northern Iran, which shared a tradition of painted pottery. These included Sialk, Cheshmeh Ali I, Late Anau I and Hissar I. He divided the periods into Sialk culture, Cheshmeh Ali culture and Hissar culture. His work relied heavily on published stratigraphy and ceramic forms, styles and decorations and provided the first integral chronology study of early Iran, based on Willard Libby's published radiocarbon dating in 1949 (Renfrew 1973).

On the other hand, the cultural sequence of Mesopotamia was used as a basis for comparison by Dyson in his synthesis of the chronology of Iran (Dyson 1965). He used the terminology of "cultural horizon" as a means of interpreting the cultural sequence of Iran. For example, in the Central Plateau sequence, various phases of Sialk I and II were considered to be related to the Hassuna, Halaf, Halaf-Ubaid, and Ubaid-Uruk Horizons of Mesopotamia (Dyson 1965, pp. 236-7, 249). Dyson also pointed out that the major problem in Iranian chronological discussions was "the tendency to rely almost exclusively upon design parallels to the exclusion of shape, non-ceramic objects, and basic technology" (Dyson 1965, p. 221). From the 1970s onwards, a number of archaeologists proposed their own chronology for the Central Plateau in which the 'types' and 'cultures' primarily were the indicators of temporal and spatial relations between different cultural groups. Negahban proposed a relative chronology for the prehistory of the Central Plateau and proposed a continuation in site occupation in the three sites of Zagheh, Ghabristan and Sagzabad, (see Supplementary Fig. A), (Negahban 1977).

Based upon the cultural-historical approach, Majidzadeh divided the Central Plateau prehistory into four distinct periods: Archaic Plateau, Early Plateau, Middle Plateau and Late Plateau, and suggested that there were two intrusive elements in the prehistory of the Central Plateau, the 'Plum Ware People' and the 'Grey-Ware' phase at Ghabristan and suggested that migration or abandonment took place in the Central Plateau between the Early to Middle Plateau periods (Majidzadeh 1981, 2008); Fazeli 2001, p. 116).

Malek Shahmirzadi, proposed four stages for the cultural sequence of the Central Plateau based on the characteristics of Ceramics: 1. Formative period, 2. Zagheh period, 3. Cheshmeh Ali period (Sialk I and II), 4. Wheel-made pottery period (or Sialk III) (Malek Shahmirzadi 1995). He also attempted to find the origins of culture on the basis of ceramic diversity. He believed that new groups migrated into the Central Plateau and instigated ceramic manufacture, then this new innovation spread throughout the region, beyond the plateau. The more recent excavations, with stricter control on stratigraphy and use of radiocarbon analyses for absolute dating, aimed to gather further information concerning the chronology and cultural development of the Central Plateau of Iran in the Late Neolithic and Chalcolithic periods. For example, the Cheshmeh Ali prehistoric site was one of the important sites that was re-excavated in 1997. Erich Schmidt was one of the prominent archaeologists who excavated the

site and distinguished two major prehistoric levels, Chalcolithic and Neolithic at Cheshmeh Ali (Schmidt, 1935 and 1936). Though, the results of Schmidt's excavation at Cheshmeh Ali were never published, however, the black on red Chalcolithic pottery unearthed by him has remained a key marker of relative chronologies for the prehistory of Iran's Central Plateau.

In the re-excavation of the site of Cheshmeh Ali in 1997 the stratigraphic sequences exhibited a pattern of change through time without any major disruption from the Late Neolithic to the Early Chalcolithic period (Fazeli 2001, p.74-76; Fazeli et al. 2004). The radiocarbon determinations taken from Tepe Cheshmeh Ali (total of 10 samples) resulted in the 5260 BC-4940 BC and 4790 BC-4540 BC dating ranges for the Late Neolithic and Transitional Chalcolithic periods, respectively (Fazeli et al. 2004).

Zagheh, a prominent site for understanding the dynamics of the Iranian plateau prehistory (see Supplementary Fig. A), was another site that was re-excavated in 2001 (Fazeli and Abbasnejad 2005). It is a low mound covering about 1.5 hectares, located at ~60 kilometres southwest of the city of Qazvin. Excavation of the site had been started by Negahban in 1970 and Malek Shahmirzadi in 1972-1973 (Malek Shahmirzadi 1979).

Sixteen domestic architectural units were identified at the site, reflecting cultural continuity throughout the site with no major or significant interruptions (Malek

Shahmirzadi 1977, p. 84).

The ceramics recovered from the aforementioned excavation at Zagheh in 2001 that were classified into four main types according to their decoration (Fazeli and Abbasnejad 2005) suggested that the site may have been a Transitional Chalcolithic period site with the co-existence of Cheshmeh Ali and Zagheh types at all levels (Fazeli and Abbasnejad 2005). Radiocarbon determinations on biological samples recovered from Tepe Zagheh were undertaken by Mashkour et al. (1999). Four C14 dates have been obtained from mammalian bone samples, and it was claimed that the settlement period for Zagheh, as indicated by these samples, ranges from 5212 to 4918 BC (Mashkour et al. 1999, p. 68).

On the other hand, the radiocarbon analysis on ten samples taken from one of the trenches of Zagheh re-excavated by Fazeli in 2001, showed that this site settled around 5370-5070 BC and was abandoned around 4460-4240 BC.

In most of the previous studies of the region, as explained above, the culture-historical viewpoint was mainly predominated and the scholars focused heavily on stylistic changes, especially the colour /decoration in ceramics.

In 2003 an international project was launched in order to study the socio-economic

transformation of the Neolithic and the Chalcolithic settlements within the Tehran plain, which later was extended to include other sites on the Central Plateau, such as the Qazvin and Kashan plains. One of the main objectives was to study the evolution of craft specialisation and settlement patterns of pre-urban societies within the Central Plateau. In this project multidisciplinary work was carried out in order to provide stratigraphic information and absolute dating using radiocarbon analyses, as well as to characterise the excavated pottery utilising petrographic study and various analytical methods. For the fulfilment of the objectives of this project three sample sites on the Central Plateau, Pardis, Ebrahimabad and Sialk, were selected which were located in the Tehran, Qazvin and Kashan plains, respectively.

Pardis site

Tepe Pardis was an important site recorded during the 2003 survey of the Tehran plain. It is located in the Tehran plain (also called Varamin plain). This is a mound of some seven metres in height above the surrounding ground level and covering an area of 4,200 square metres. Fazeli et al. (2010) by conducting a preliminary study on collections of well-stratified potsherds excavated in the Tepe Pardis site reported a series of important technological changes in ceramic production from the soft, buff vessels of the Late Neolithic period to the distinctively harder red and black-painted wares of the Transitional Chalcolithic period. According to them this change did not involve different raw materials or higher firing temperatures, but probably required

longer times of firing and a more efficient and ingenious control of the firing process. This evolution was related to the invention of sophisticated four-chambered kilns discovered in rows in this specialised ceramic-producing village.

Sialk site

Sialk, one of the most important sites of this region for exhibiting a nearly continuous archaeological sequence from the sixth millennia BC, is located in the Central Plateau of Iran, southwest of Kashan city and consisted of two, north and south, mounds which are ~ 600 metres apart. Roman Ghirshman, who was one of the first archaeologists conducting systematic archaeological investigations on the Central Iranian Plateau, excavated the Sialk site for the first time. He demonstrated that there was a gradual development at the site from the Late Neolithic period, with cultural continuity demonstrated through ceramics and architecture (*ibid.*). Ghirshman opened three large trenches (I, II and III) on the North Mound (Ghirshman, 1938) and divided it chronologically into two main phases, Sialk I and Sialk II. The lowest level of the North Mound, called Sialk I (ca. 6000-5200 BC, Late Neolithic period) mostly contained the pottery possessing coarse buff body with black painted decoration (Ghirshman, 1938, p. 11f.) and Sialk II (ca. 5200-4600 BC, Transitional Chalcolithic) represented the upper part of Sialk North Mound, comprised of red pottery, painted in black (Fig.1). Ghirshman also proposed five sub-phases for Sialk I (II1-II5) and three for Sialk II (III1-III3) based on similarity of pottery design. The North Mound was also

re-excavated by Fazeli and Coningham, in 2008 and 2009. In this project, which aimed to provide absolute dates for the earliest occupation levels at the site, one stratigraphic stepped trench was excavated into the west section of Ghirshman's original trench II. This trench, measuring 2.5 by 2 metres, yielded virgin soil to a depth of 16.15 metres. The complete sequence comprised of 152 layers, including several occupational floors, pits and remains of walls (Fazeli et al. 2013). (See the chronology of the site in Supplementary Table. A).

The two major types of Sialk pottery, i.e. Sialk I and II, excavated from the North Mound are regionally distributed within the whole Central Iranian Plateau and the prehistoric chronology of the Central Plateau has been based almost entirely on these types of pottery. In the present study the pottery recovered from Sialk has been analysed and characterised utilising various scientific techniques.

AIMS AND OBJECTIVES

In the present study the Pottery samples from the North Mound of Sialk, as excavated by Fazeli and Coningham (2008-2009), were investigated in order to determine their chemical and mineralogical compositions and microstructures to bridge the gap in our knowledge regarding the technical aspects of pottery making and its development at Sialk. In most of the previous excavations, as explained above, the colour

/decoration has been the main criterion used for identification, characterisation, and comparison of various pottery of the region and they have never been studied in terms of technology of production, as rightly stressed by Dyson (Dyson 1965, p. 221).

In this way pottery of similar colour and decoration has usually been classified and named with a single common name, (sometimes called tradition) e.g. Sialk I or II. Hence, the exact origin of similarities between different pottery (export of products, cultural interaction or technology transfer) could never be discovered.

This could also result in some confusion and misunderstanding regarding the exact nature of socio-economic exchanges between various prehistoric societies, e.g. such as the assumption of existence of an intrusive element from outside that brought about the changes in societies, abandonment of settlements in some areas of the Central Plateau (Majidzadeh 1981, 2008), or migration of some people into the Central Plateau who imported the ceramic manufacture to the region (Malek 1995). These propositions were simply based on the finding of some pottery with apparently novel and different colour/decoration in comparison with the existing ones.

The results of the present study would provide the material basis for investigating the nature of changes in the ceramic production techniques which occurred during the Late Neolithic to Transitional Chalcolithic transition period

(ca. 5200-4600 BC) in this site. This also would introduce a more reliable criterion for comparison of the Sialk pottery with other pottery of the Central Iranian Plateau and to clarify the nature of existing interactions between Sialk and other prehistoric communities of the region.

This study in particular, will try to address the following questions:

- What caused the general chromatic change of the pottery from the Late Neolithic to the Transitional Chalcolithic periods?**
- Did this change involve a replacement of selected base materials or rather was it a consequence of refinement of the firing technology?**
- What evidence is there for continuity, change in ceramic technology, long distance trade, and other evidences of mass production and specialization in making ceramic wares at the site witnessing the Late Neolithic to the Transitional Chalcolithic transition?**
- Is there any evidences for the interference of foreign elements in the**

change of production of pottery, or importing the ceramic manufacture to the site?

To the authors' knowledge, the techniques utilised in the present work have never been used to characterise the prehistoric pottery of this site.

EXPERIMENTAL PROCEDURE

Sample selection

In this study 36 samples comprised of 22 and 14 sherds of the Sialk I and II type, respectively, were selected from the collection of C14-dated pottery sherds which were recovered during the two fieldwork seasons conducted by Fazeli and Coningham in the North Mound of Sialk in 2008-2009. The Sialk I and II samples were selected randomly from earlier to later sherds (according to their chronological information) among two separate pottery collections each containing one type of the aforementioned pottery.

Chemical analysis

There are many different techniques that can and have been used for chemical

analysis of archaeological artefacts. In this research we used X-ray fluorescence (XRF) technique, utilising an Oxford ED2000 spectrometer.

The XRF spectrometer is an x-ray instrument used for routine, relatively non-destructive chemical analyses of rocks, minerals, sediments and fluids. XRF analysis is useful for investigating around 80 elements present in major quantities (Rice 2005, p. 394; Pollard et al. 2007 p. 101).

Mineralogical analysis

The study of the thermal behaviour of ancient pottery has always attracted much attention in archaeological sciences because it yields useful information about the technology of making and firing of ancient pottery. X-ray Diffraction (XRD) is one of the most popular techniques for identifying the minerals present in ceramics (Rice 2005, pp. 382-386; Pollard et al. 2007 p.103). Detecting and determining the nature of the minerals present in the ceramics by XRD could be used as an indication of the firing history of the bodies. The estimation of firing temperature by XRD is based on the fact that the mineralogical composition of clays changes during firing. These changes normally comprise of the loss of water from the clay minerals and other hydroxides, the decomposition of the carbonates with loss of CO₂ and the formation of various new phases and crystalline minerals. The changes which occur during firing of clay bodies can be monitored by XRD. Considering this fact, XRD, as a direct method, has constantly been of major interest in determining firing temperature

of ancient ceramics (Holakooei et al. 2014).

In this study 20 sherds from Sialk (10 samples each from Sialk I and II) were selected randomly and were analysed by Powder X-ray Diffraction analysis (PANalytical X'pert Multi-Purpose Diffractometer), using Cu K α 1 radiation and a PIXcel solid-state detector in a 2θ range of 5–120°. The step-scan size was approximately 0.013° in 2θ and the total acquisition time per pattern was 40 minutes. Identification of crystalline phases by XRD was carried out using the International Centre for Diffraction Data Powder Diffraction Files (ICDD PDF). The JCPDS reference cards were used to interpret the patterns.

Microstructural examinations

Scanning electron microscopy (SEM) analyses the surface of materials and provides detailed high-resolution images of the sample by rastering a focussed electron beam across the surface and detecting secondary or backscattered electron signals. An Energy Dispersive X-ray Analyser (EDX or EDA) is also used to provide elemental identification and quantitative compositional information (Pollard et al. 2007, p. 109).

In this research, in some cases, X-ray mapping of specimens was also created. In this method the whole surface area analysed is systematically mapped in terms of mineralogy or elemental composition and the resultant data provides a false colour mineralogical/compositional map of the sample. In addition, the compositional data is

reported as modal mineralogy in area %, along with the size of each discrete mineralogical component.

Sometimes the nature and extent of the changes which occurred during the firing process in ceramics, such as the estimation of the degree of vitrification within the clay matrix of ceramics can also be observed and determined by SEM.

In this study some typical samples were subjected to SEM examination (Hitachi TM-3000) and phase compositions of certain zones in the microstructures were determined by an EDX (Swift ED) attached to the SEM. For this kind of SEM analysis no preparation of samples was needed.

RESULTS AND DISCUSSION

Chemical compositions

Table 1 lists the chemical compositions of some Sialk I and II specimens. Obviously, the Sialk I specimens exhibit quite similar compositions. The relatively high value of standard deviation shown by CaO is common and can be attributed to the variation of its content in the original clay deposits. It has been shown that of six elemental oxides, the greatest variation within a single clay deposit occurs with CaO (Buko 1984). In the burial environment, a variety of processes may also alter the chemical composition of pottery. Two of these processes are cation leaching (Bieber et al.

1976) and exchange (Hedges and McLellan 1976). Calcium is one of the elements that are susceptible to all of the above-mentioned processes. Leaching and ion-exchange processes may also affect the alkali elements such as sodium and potassium, whereas silicon, aluminium and iron are more resistant to these processes. On the basis of these results, it may be deduced that the pottery studied herein has been made using a single resource of clay raw material, or clay from very similar resources and the relatively high content of CaO in most specimens indicates the use of calcareous clays as the source of raw material to make most of this pottery.

On the other hand, the chemical composition of the Sialk II type specimens indicated the existence of two different types of pottery, calcium rich and relatively poor in calcium, each group exhibiting quite homogeneous compositions. The group one specimens (calcium rich), which have almost similar compositions to the Sialk I specimens, apparently have been made using the same clay raw materials as the Sialk I pottery. These vessels are distinguished by the strong red colour of their surface and buff colour of the core, while the specimens of the second group, which were red both in the core and on the surface (as shown in Table1) are the product of different raw materials.

Mineralogical analysis

Table 2 summarises the mineralogical analyses of Sialk I and II specimens (for PXRD traces see Supplementary Fig. B). It can be seen that besides the signs of the presence of CaCO_3 in some specimens, especially the older specimens, quartz and esseneite were the major crystalline phases of Sialk I and calcium rich Sialk II specimens, whereas, the low calcium Sialk II specimens were mainly composed of quartz, hematite and augite phases. Moreover, the faint trace of illite phase observed in some specimens (which is more pronounced in older specimens of Sialk I pottery e.g. specimen S1z), indicates that the raw materials used in production of this pottery, except the low calcium Sialk II pottery, has possibly been illite clays.

Microstructures

Figures 2 show the typical SEM micrographs of (a) Sialk I and (b) Sialk II, respectively. The relatively uniform microstructures and the absence of large and angular particles indicate that the raw materials used were most probably sedimentary clays, and that no inorganic tempers have deliberately been added to the starting clays. However the traces of plant tempers, such as fine chaff or dung, can be seen on the cross sections of both pottery samples.

According to electron microscopy, while, each group of the Sialk I and II types of pottery exhibited quite homogeneous microstructures within themselves, a marked differences related to density, degree of vitrification and porosity were observed between the two different groups of pottery, for example, the earlier Sialk I sample,

S1h, and the latest Sialk II sample, S2k, as shown in Figs. 2a and 2b, respectively.

The latter pottery sample which is red both on the external surface and at the core, exhibits more dense, vitrified and pore-free microstructure (Fig. 2b) in comparison with both the S1h pottery sample (Fig. 2a) and even the Sialk II pottery with red external surface and buff core (for example see Supplementary Fig. C).

This difference in structural characteristics, obviously, would affect some of the crucial properties of the pottery, such as the mechanical strength and permeability.

Effect of heat on pottery and estimation of the firing temperature

According to Rice (1987, p. 98) and El-Didamony et al. (1998) CaCO_3 decomposes at 800–850 °C and the lattice structure of the illite clays is collapsed in the 850-1000 °C temperature range (Rice 1987, p. 92). In calcareous clays fired to 850 °C or above, the presence of CaO may cause some problems, since calcium oxide particles are highly hygroscopic. Over time, they may pick up moisture, forming quicklime, which is accompanied by volume expansion and stresses, causing cracking and spalling of the surrounding clay body (Rye 1976). If the CaO content is high and the particles are relatively large, this may give the fired ware a low strength. If the body is fired at temperatures 850-900 °C or beyond the rehydration does not occur, since at these temperatures calcium in clays becomes part of a liquid phase, with sintering and vitrification. Tite and Maniatis (1975a) suggested that in calcium and magnesium rich clays melting begins at lower temperatures (approximately at 800 °C), because Ca and

Mg may act as fluxes (Segnit and Anderson, 1972). That is why ancient potters might have explored these kinds of clays to make their pots by using less energy.

Didamony et al. (1998) studying the firing behaviour of calcareous clays also observed distinct firing shrinkages in the 1050-1150 °C temperature range, which were attributed to the formation of a liquid phase in compositions located in the vicinity of the major eutectic of the SiO₂–CaO–Al₂O₃ system. This eutectic has CaO/SiO₂ and Al₂O₃/SiO₂ molar ratios of 0.402 and 0.140, respectively, and a fusion point of 1165 °C. Obviously, the most efficient densification and vitrification process should occur in the above temperature range. The Sialk I pottery samples, owing to their lower Al₂O₃/SiO₂ ratio and higher ratio of CaO/SiO₂ can be expected to show lower sintering and vitrification temperatures than the 1050-1150 °C range reported by the above researchers. On the other hand, the Sialk II sherds are more refractory because of their much lower Al₂O₃/SiO₂ ratio and their sintering and vitrification could occur at higher temperatures in comparison to the Sialk I pottery. It has also been reported that new calcium compounds, such as diopside, wollastonite and calcium ferrosilicates, are mainly formed at high temperatures 900-1100 °C (Tite and Maniatis 1975b). On the basis of PXRD analysis and microstructural studies, it can be claimed that apart from very few CaO particles present in some specimens, which are the remnants of large particles that did not have enough time to react during firing of the pottery, there is no evidence for the presence of CaO particles in the specimens. On the other hand, there is the clear sign of formation of calcium, iron, aluminium silicate minerals like esseneite in the Sialk I pottery. Moreover, it can be

deduced that the iron oxide liberated from the decomposition of some clay minerals, such as illite, has possibly been incorporated in the silicate lattice structures at temperatures lower than shown in the above reports. Considering the fact that the main silicate mineral present in the Sialk I specimens is an esseneite like mineral, and that no hematite crystals were detected in the sherds specimens, it can be deduced that iron has mainly been incorporated into the esseneite crystal structure, which could accommodate high amounts of iron. Hence, considering the decomposition and distraction temperatures of calcium carbonate and clay minerals, the sintering and vitrification processes and the formation temperatures of the silicate and aluminosilicate phases it can be suggested that the Sialk I pottery has possibly been fired at the 900-950 °C temperature range, except the older pottery (e.g. specimen S1z, Table 1) that has been fired at lower temperatures ~800-850 °C.

The two different types of Sialk II pottery, calcium rich and poor in calcium, showed different microstructures and phase compositions. The group one specimens (calcium rich) are distinguished by the strong red colour of their surface and buff colour of the core, while the specimens of the second group were of red colour both in the core and on the surface. SEM elemental map (Fig. 3) showed that the fine red slip present on the interior and outer surface of the latter Sialk II sherds contained pigments rich in iron oxide. The aforementioned sherds, except the red coating, possessed similar phases as the sherds of Sialk I pottery, namely quartz and esseneite. It is interesting to note that there are also a few Sialk I sherds that were covered with a red coating (the

last four samples of Sialk I samples shown on Table 1). Hence, it can be concluded that the technique of applying red coatings on the Sialk pottery had been an old technique that continued from Sialk I to Sialk II periods. On the other hand, hematite was identified in all the samples of the second group of Sialk II specimens, which belong to the later phases of this period. Although the iron oxide minerals may be present in the clays used as raw material in the production of pottery, this oxide can also be generated during firing of pottery in an oxidising atmosphere, as the product of destruction of iron containing minerals present in raw materials and recrystallisation of secondary hematite crystals. However, it must be taken into account that calcareous lumps in calcium-rich clay may prevent the formation of hematite crystals by fixation of iron in the network of newly-formed calcareous silicate and aluminosilicate minerals and, consequently, inhibit the generation of red colour in the fired pottery (Rice, 1987, p.336).

The aforementioned process has apparently occurred in the case of Sialk I pottery, as discussed above, whereas in the case of the Sialk II, bulk red pottery, which appeared at the later phases of the period, the major calcium aluminium iron magnesium silicate mineral (augite) accommodates much lower amounts of iron oxide. Moreover, owing to the low content of calcium in the clay raw materials, the volume of the augite mineral would be lower in comparison to the esseneite mineral in Sialk I pottery. Hence, a great proportion of iron oxide present in the raw materials of this group appeared as the iron oxide mineral (hematite) in the fired bodies.

The Sialk II, bulk red pottery sherds studied in this work exhibited quite dense and vitrified microstructures, (Fig. 2b). This could only have been achieved by the presence of a liquid phase, in a sufficient quantity and with a relatively low viscosity, at the maximum firing temperature. Considering the higher refractoriness of the raw materials used in the production of the above pottery, as discussed above, a higher firing temperature should be anticipated for this pottery in comparison to the above-discussed bodies, including the Sialk I potter. Therefore, on the basis of the above facts and observations the temperatures range of 1050-1100 °C can be assigned to the firing of the low calcium Sialk II pottery investigated in this work. It should be noted that the other types of Sialk II-type pottery, having red coating owing to their similarity in chemical and mineralogical compositions to the Sialk I (buff) pottery of the same site, probably have been fired at nearly the same temperature range of the Sialk I pottery. It should be noted that in non-industrial firing there might be considerable fluctuations in firing temperatures. Even in kiln firing, temperature differences of as much as 100 °C may exist between different sections of the kiln (Mayes 1961, 1962). Under these conditions, the determination of the exact firing temperatures is impossible, but the quite high range of firing temperatures, as stated above, and the good quality of the fired ware (very dense products with no deformation) indicate that the early potters of the region had remarkable skill and experience in the selection of raw materials and shaping and firing techniques of pottery.

The course of evolution in the pottery making at Sialk

In the basis of the archaeological data obtained from the excavations at Sialk and the experimental results discussed above, it can be deduced that a gradual development at the site can be observed from the Late Neolithic, with cultural and technological continuity demonstrated through the gradual evolution of the pottery-making industry. This is in accordance with the findings of Ghirshman (1938), who demonstrated that “there was a gradual development at Sialk site from the Late Neolithic, with cultural continuity demonstrated through ceramics and architecture”.

According to the C14-dated sequence of the Sialk site, as shown in Tables 3, the pottery of the main groups of Sialk I and II can be divided into four sub-groups of a, b, c and d according to their dating. The sub-group (a) the earliest pottery sherds collected from the contexts 6032 and 6035 (dated as 5894-5725 BC) as well as 6036 and 6042 (dated as 5775-5642 BC), and the intermediate sub-group (b) collected from contexts 6013 (dated as 5465-5442 BC), and the sub-group (c) collected from contexts 6018, as well as 5117 and 5095 (dated as approximately 5300-5200 BC range), and the latest sub-group (d) collected from contexts 5026, 5021 and 5017 (dated as 4982-4973 BC). The first three sub-groups mostly belong to the Sialk I buff pottery group, which, as discussed above, exhibit little difference in chemical and mineralogical composition as well as microstructure. For example, the samples S1ae and S1c, from the sub-groups (a) and (c), respectively, in spite of a difference of more than 500 years in dating,

show similar mineralogical compositions and microstructures. It is interesting to note that some of the pottery of the Sialk I group (as specified by their decoration) belonging to the latest sub-group of (c) such as the samples S1ac, S1ad and S1r contained a red coating on their interior and exterior surfaces. This indicates that in the later stages of Sialk I period the application of red coating on the pottery surfaces has been practiced by the potters of Sialk and this tradition continued from Sialk I to Sialk II periods. This finding also implies that the red colour and the specific decoration of the Sialk II pottery are not necessarily coincidental with each other.

On the other hand, the latest subgroup (d) contained the two different types of red Sialk II pottery, calcium rich and poor in calcium. The calcium rich specimens have been covered with a red coating, whereas, the specimens of the other group (poor in calcium) were of red colour both in the core and on the surface.

As discussed above, the aforementioned group of Sialk II specimens showed substantially different microstructures and phase compositions. The pottery sherds covered with a coating, except the red coating, possessed similar phases as the sherds of Sialk I pottery, namely quartz and esseneite, whereas, the other group of pottery sherds, which were red both in terms of surface and core, mainly contained quartz, hematite and augite (Calcium Aluminum Iron Magnesium Silicate) phases.

On the basis of the above findings it can be proposed that an apparent gradual development at the site of Sialk from the Late Neolithic through to the Transitional Chalcolithic periods, (from ~ 5900 to 5000 BC) can be observed. However, the course of the development in the pottery industry at first was very sluggish. In fact it seems that there is no substantial difference between the pottery quality and structure (as proved by the mineralogical composition and microstructural study) from the beginning of the Sialk I period to the end of red-coated Sialk II pottery. During this period, in fact, no distinct change has occurred in the process of making pottery, in connection to resources and firing technology.

However, the pottery industry witnessed a very distinct change from the Sialk I to Sialk II period by producing bulk red pottery. It is interesting to note that this change didn't necessarily coincide with the fundamental alteration of the pottery decoration, since, the use of the new decoration known as the Sialk II, (black painted motifs, consisting of simple or composite geometric designs), began in the early Transitional Chalcolithic period with the production of some apparently red pottery, which in fact was the previous Sialk I-type buff pottery covered with a red coating. This change in decoration apparently did not involve substantial alteration in technology, other than exerting more control on the atmosphere of kilns (which of course was an advancement of firing technology) which is not comparable to the radical changes in technology concerning the

selection of raw materials and the mastering of firing techniques, i.e. the careful control of the firing temperature and atmosphere, essentials of the production of bulk red pottery, that was realised in the later stages of the Sialk II period.

The production of the latter pottery is an event that can be evaluated as a breakthrough in the process of evolution of the pottery making in Sialk. It is suggested that the commencement of the production of the aforementioned high quality pottery should be considered as the critical time of entrance into the transitional chalcolithic era.

This suggestion, though is a confirmation of the idea of Ghirshman (1938) regarding the observation of a gradual development at the site of Sialk from the Late Neolithic period and also is in accordance with Malek Shahmirzadi's (1995) suggestion that Period I and II should be considered as one, since, many of the features of Period I continue into Period II, as well as Wong's (2008) proposal that the ceramic industry of Period II is essentially a continuation of Period I (which is characterised by an improvement of firing conditions). **However, it should be noted that none of the previous researchers recognised the prominence of the aforementioned production phase and didn't differentiate the two types of Sialk II red pottery, fundamentally differing in technology and quality of the ware.**

As discussed before, Fazeli et al. (2010) also reported changes from the apparently softer, buff painted vessels of the Late Neolithic to the production of distinctively

harder red and black-painted wares of the Transitional Chalcolithic period in the Pardis site, which according to the authors did not involve different raw materials or higher firing temperatures, but only required longer times of firing and a more efficient control of the firing process. This is contrary to our findings at Sialk, indicating that the production of the **bulk red ware** usually necessitated the use of low calcium clays (differing from the calcium rich clays used in production of the buff pottery) and higher firing temperatures and more oxidising atmospheres. It is very difficult to explain the cultural and socio-economic causes behind the adaption of different raw materials and firing techniques (perhaps more sophisticated kilns) for production of the bulk red pottery. While the change of decoration is quite common amongst the prehistoric pottery makers, and usually can be attributed to the cultural connections and interactions of the prehistoric communities, the change in raw material resources and firing techniques often encountered strong resistance, hence, were less common. However, it has been discovered that many features of the pottery making tradition in Sialk, such as the form and decoration of pottery, has been preserved during the gradual evolution of pottery making at Sialk, commencing from the coarse, fragile buff pottery to the stronger bodies covered with a red slip and eventually leading to the high quality, fine and quite strong, bulk red pottery. Obviously, **this development could only have been realised by the gradual progress in the skill, knowledge and information of the local potters, rather than the occurrence of some abrupt socio-economic changes, or outside interference.**

Moreover, it should be stated that the efficient control of the temperature and time, i.e. maintaining the required degree of oxidising atmosphere and observation of the appropriate time-temperature schedule during the firing of pottery, which are the main conditions for producing bulk red pottery, could also result in the elevation of the firing temperature and the occurrence of more efficient sintering and vitrification processes. This would give rise to denser and stronger red pottery (Fig. 2b), **hence, the production of bulk red pottery, which is of red colour both in the core and on the surface, in addition to the possible aesthetic considerations, are of prime importance in producing more dense impermeable and strong pottery.**

Considering the quantity and quality of the pottery recovered from the site, the continuation and gradual development of pottery production and the similarity of the products belonging to each period regarding their form, decoration as well as their chemical/ mineralogical compositions and microstructures seems that some fundamental conditions accompanying the transition of the Late Neolithic to the Transitional Chalcolithic period within the communities of the Iranian Central Plateau have been realised at Sialk, at the time of occurrence of the aforementioned change. These conditions are the establishment of specialised craft areas, standardisation of craft production and using of kilns for ceramic production (Fazeli et al 2007). Though no kilns were discovered during the excavations in Sialk (perhaps because of the limited extent of the excavations carried out on the site) the aforementioned strict control exerted on the firing atmosphere and temperature, as well as the observation of the careful time-temperature relation during the firing process of pottery at the later

stages of the Sialk II period, as stated above, would have been impossible without the use of relatively sophisticated kilns in Sialk in that specific period.

CONCLUSIONS

Chemical and mineralogical analyses along with microstructural studies of Sialk pottery showed the quite homogeneous nature of the sherds, both chemically and mineralogically, and revealed the occurrence of a gradual evolution and development in pottery making at the site commencing from the coarse, fragile buff pottery to the stronger bodies covered with a red slip and eventually leading to the high quality, fine and quite strong, bulk red pottery in the later stage of the Sialk II period. The aforementioned latter stage of Sialk II is an event that can be evaluated as a breakthrough in the process of evolution of the pottery making in Sialk, and should be considered as the critical time of entrance into the Transitional Chalcolithic era.

It was also shown that the chemical compounds present in the sherds were the products of reactions occurring at temperatures of 900-950 °C and 1050–1100 °C for Sialk I and II, respectively. These results, revealed the existence of a high degree of skill and experience in the potters of Sialk, at the later phase of the Sialk II period, concerning the techniques of selection of raw materials and mastering of the firing techniques. Without the existence of this high degree of specialisation in the pottery making in Sialk II, developed over several centuries in this region, the production of the fine, well-made and dense red pottery could not have been achieved at the aforementioned time in this site.

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FIGURE CAPTIONS

Figure 1. Sialk pottery sherds: (a) Sialk I Buff Ware, (b) Sialk II Red Ware.

Figure 2. SEM microstructures of the Sialk pottery sherds: (a) Sample S1h, (b) Sample S2k. The black particles show the added organic temper to the pottery.

Figure 3. SEM microstructure showing the surface of a Sialk II specimen covered with a red coating rich in iron oxide. Mixed map: Calcium (red), silicon (green), iron (blue).

TABLE CAPTIONS

Table 1. The chemical compositions of some Sialk I and II pottery samples (wt%).

Table 2. Major crystalline phases present in some typical pottery samples.

Table 3. C14 dated sequences of some Sialk I and II pottery samples.

SUPPLEMENTARY MATERIAL CAPTION

Supplementary Figure A. A map showing the location of Sialk and some of the other prominent prehistoric sites in the Central Plateau of Iran.

Supplementary Figure B. XRD traces of some typical specimens.

Supplementary Figure C. SEM microstructure of the red coated Sample S2n, (core).

Supplementary Table A. Chronology of the Central Plateau of Iran based upon evidence from the excavated sites (After Fazeli et al. 2009).

SUPPORTING INFORMATION

Additional Supporting Information may be found in the online version of this article at the publisher's web site:

Supplementary Figure A

Supplementary Figure B

Supplementary Figure C

Supplementary Table A